TITLE: Acceleration of Field-scale Bioreduction of U(VI) in a Shallow Alluvial Aquifer: Temporal and Spatial Evolution of Biogeochemistry

PRINCIPAL INVESTIGATOR:

Philip E. Long Phone: 509 372-6090 FAX: 509 372-6089

Pacific Northwest National Laboratory

Mail Stop K9-33; P.O. Box 999, Richland, WA 99352 E-mail:philip.long@pnl.gov

COLLABORATORS: Derek R. Lovley¹, Kelly Nevin¹, Regina Wilson¹, C. T. Resch², Aaron Peacock³, Helen Vrionis¹, Yun-Juan Chang³, Dick Dayvault⁴, Irene Ortiz-Bernad¹, Ken Williams⁵, Susan Hubbard⁵, Steve Yabusaki², Yilin Fang², and D. C. White²

¹University of Massachusetts, Amherst, MA; ²Pacific Northwest National Laboratory, Richland, WA; ³University of Tennessee, Knoxville, TN; ⁴S. M. Stoller Corporation, U.S. Department of Energy, Grand Junction, CO; ⁵Lawrence Berkeley National Laboratory, Berkeley, CA.

Uranium contamination in a shallow aquifer at Rifle, CO provides opportunity for repeated field-scale electron donor amendment experiments allowing investigation of rates of U(VI) reduction under varying terminal electron accepting processes. In the first two experiments conducted in the same experimental plot during the 2002 and 2003 field seasons, U(VI) loss from groundwater occurred synchronously with growth of Geobacter after amendment of the subsurface with acetate at concentrations of either ~3 mM (2002) or ~10 mM (2003). These two experiments demonstrated the importance of 1) maintaining Fe(III) reducing conditions for optimal U(VI) bioreduction and 2) controlling the spatial location of sulfate reduction to facilitate downgradient Fe(III) reduction. Monitoring of U(VI) concentrations and other geochemical parameters after the 2003 experiment suggests that loss of U(VI) as groundwater flows through the test plot can be maintained for >1 year after last addition of acetate, particularly in zones where high levels of bioreduction occurred during acetate amendment. Mechanisms for removal of U(VI) from groundwater in the absence of acetate amendment are not known and will be a focus of future field-scale experiments.

During the 2004 field season, acetate was amended to the subsurface in a new, smaller experimental plot under conditions similar to that of the 2002 experiment. The objectives of the 2004 experiment included 1) Determine if the U(VI) loss and Geobacter growth of the 2002 experiment can be replicated at a second location, 2) Define the growth of Geobacter with high frequency sampling during the first month of acetate amendment, 3) Collect and filter groundwater samples for mRNA and genomic analysis under biostimulated conditions, and 4) obtain preliminary geophysical measurements (complex resistivity) to assess the possibility of using non-invasive techniques to monitor microbe-mediated changes in subsurface mineralogy. Analysis of data from the 2004 experiment is in progress, but preliminary results indicate that all objectives of the experiment were met. Uranium concentrations in groundwater down gradient decreased in a fashion similar to the 2002 experiment except that the experiment was terminated prior to development of extensive sulfate reduction. The decrease in U(VI) paralleled dominance of Geobacter in groundwater samples, strongly indicating that Geobacter is responsible for enzymatic reduction of U(VI) in situ (see Lovley et al. abstract for clone library data and mRNA results). Inversion of complex resistivity data show a marked anomaly below the water table in the vicinity of the injection gallery which is interpreted as a response to the changed redox status of Fe-bearing mineral coatings on detrital grains. This promising result suggests that more extensive resistivity measurements may provide detailed tracking of Fe redox status in response to in situ bioreduction. Future research at the Rifle site will include detailed reactive transport modeling and additional in situ experiments that will elucidate controls on U(VI) reduction and reoxidation rates under both Fe reducing and sulfate reducing conditions.